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Sticking of He^4 on cold NaF and LiF surfaces in presence of resonant bound states

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Abstract: One phonon assisted stickings of He^4 on LiF and NaF are considered quantum mechanically under T-matrix formalism. The bound state energies and corresponding surface temperatures at which the sticking takes place are evaluated and compared with the experimental results.It is found that the exact T-matrix approach using the Morse potential gives better results.

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Key words : Phonon assisted Sticking, Bound state, Life time, T-matrix.

The mechanism of sticking of light atoms and molecules on the cold solid surfaces are actually tackled by quantum mechanical approach as the results give the better agreement with experiments on molecular beam.The probability of transition from an initial continuum to a negative energy

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bound-state(BS) can be interpreted as the trapping or the sticking coefficient(SC) for a given BS.We intend to calculate the SC in presence of resonance BS.

In addition to formation of BS the inelestic processes become very important and responsible for sticking when we choose the low energy incoming particles. This is interpreted as the phonon assisted scattering of a gas solid interaction and normally treated by distorted wave Born approximation(DWBA).

For phonon assisted inelestic scattering leading to possible physisorption, the quantum reflection effects which reduce the adsorption coefficient to zero in a limit of zero energy could very well be destroyed by successive phonon interaction.The usual method of DWBA may not be adequate as it overcounts the scattering channels.[1]

We consider this as our starting point and conjecture that this may be taken care of by considering the proper corrections to inelastic scattering.The purpose of the present work is therefore to evaluate the sticking in presence of BS[2] taking the higher order terms in the scattering T-matrix of the system. For phonon assisted adsoption in which the one phonon is taking part,the controlled adsorbtion process is completed after an equilibrium adsorbate has been built up with the gas temperature T_g equal to the initial solid temperature and then suddenly lowered by T_s[3].In evaluating the proposed parameters we assume the following:

- The coupling is restricted to be linear in the displacement and the scattering is considered in low energy region(for one phonon)
- The particles are assumed to be structureless and surface to be flat in the rigid lattice situations.
- The interaction potential has been chosen to be Morse-potential which develops just one shallow BS ($|E_0| \leq \hbar\omega_D$).
- During adsorbtion process no chemical reaction takes place.
- The spectral distributions for the surfaces have been assumed to be the Morse type and the spectral functions are obtained from the Green's functions of adsorbed surfaces.

The model Hamiltonian for the gas-solid system can be separated into the following sub Hamiltonian for gas,solid and the interaction part i,e

$$H = H_g + H_s + H_{dyn}, \tag{1}$$

Which in terms of creation/annihilation operators and in localised, non-localised, phonon modes and after canonical transformation reduces to[4],

$$H = \sum_q E_q c_q^\dagger c_q + \sum_p \hbar\omega_p b_p^\dagger b_p + \frac{1}{2}\sum_{q,q'} \chi(q-p,q)\chi(q'+p,q')\lambda_p^2 c_{q'+p}^\dagger c_{q'} c_{q-p}^\dagger c_q \times [Y_{-p} - Y_{+p}], \tag{2}$$

Where $Y_{-p}=[\frac{1}{(E_q-E_{q-p}-\hbar\omega_p)}]$ and $Y_{+p}=[\frac{1}{E_{q'}-E_{q'+p}+\hbar\omega_p}]$

In equation (2) the first part represents the localised free particle energy where E_q is the eigen value of the free particle state containing the BS energy E_n with n=0,1,2,3,.....The second part represents the kinetic energy pf the harmonic solid with $b_p^\dagger$ b_p the creation/annihilation operators of longitudinal acoustic phonons of frequency ω_p in the absence in gas ($\lambda_p^2 = \frac{1}{\omega_p}$) with phonon modes ω_p. The third term represents the gas solid interaction with kernel given by

$$\chi(q,q-p) = (\frac{\hbar}{2M_s N_s})^{-1/2} \int \phi_{q-p}^\star(x)\frac{dV_0(x)}{dx}\phi_q(x)dx. \tag{3}$$

For just one shallow BS we take the static surface potential as,

$$V_0(x) = U_0(e^{-2\gamma(x-x_0)} - 2e^{-\gamma(x-x_0)}). \tag{4}$$

To obtain the T-matrix we solve the Hamiltonian with the localised single particle Green's function

$$G_{km}(t) = << \alpha_k(t), \alpha_m^{\dagger}(0) >> . \tag{5}$$

Which after Fourior transformation and iteration leads to

$$G_{kk}(E) = G_0(E) + G_0(E)TG_0(E), \tag{6}$$

where,

$$G_0(E) = \frac{1}{2\pi(E - E_k)}; \; T = \frac{2\pi\Delta_k}{1 - \sum_q \frac{\Delta_q}{(E-E_q)}}$$

$$and \quad \Delta_q = \sum_p \frac{|\chi(q, q-p)|^2 \lambda_p^2 n_{q-p}}{E_q - E_{q-p} - \hbar\omega_p}. \tag{7}$$

Here 'q' is the momentum of the gas particle in the localised state and $(q - p)$ is that in the BS. E is the effective final energy with transformed BS energy $E_{n'}$ due to gas solid interaction plus the phonon energy.

Hence the transition probability from the initial continuum to final BS under the emission of phonon may be given by the Golden rule

$$R_{cn} = \frac{2\pi}{\hbar} < \psi_f | Re \; T \; |^2 \psi_i > \delta(E_f - E_i) \; n_{eq}. \tag{8}$$

where for BS energy E_i,

$$n_{eq} = exp(\beta_g \mu - \beta_s E_i)$$

The real part of the T-matrix gives the transition probability R_{cn} and hence the sticking coefficient normalised by the flux of incoming particle τ_L defined by $\tau_L = \frac{2Lm}{\hbar k}$ which is

$$S = \frac{h^2\gamma^2}{4mk_\beta T_g} \left[\frac{C}{\int_0^\infty F(x)dx}\right]^2 n_{eq}. \tag{9}$$

where,$C = F(s^2)$ and

$$F(x) = \frac{\sinh(2\pi\sqrt{x})}{\sinh^2(\pi\sqrt{x}) + cosh^2(\pi\sigma_0)} \left|\Gamma(1/2 + \sigma_0 + i\sqrt{x})\right|^2$$
$$\left[x + (\sigma_0 - n - 1/2)^2\right]^2 \left(\frac{x - s_n^2 + s^2}{s^2}\right), \qquad (10)$$

with $n_{eq} = exp\,|\beta_g \mu|\, exp[-\delta s^2/r]$ where $\delta = \beta_s \hbar\omega_d$, $\sigma_0^2 = \frac{2mU_0}{\hbar^2\gamma^2}$; $S_n^2 = \frac{2m|E_n|}{\hbar^2\gamma^2}$; $r = \frac{2m\omega_d}{\hbar\gamma^2}$

TABLE(1): Morse-Potential parameters[5] for He-LiF and He-NaF systems.

systems	$\gamma^{-1}(A^0)$	$U_0(K)$	$\frac{\hbar\omega_d}{K_\beta}(K)$	σ_0	r	$\frac{m}{M_s}$
He-LiF	1.09	81.75	730	4.023	144.55	0.152
He-NaF	0.97	77.78	450	3.491	70.496	0.0952

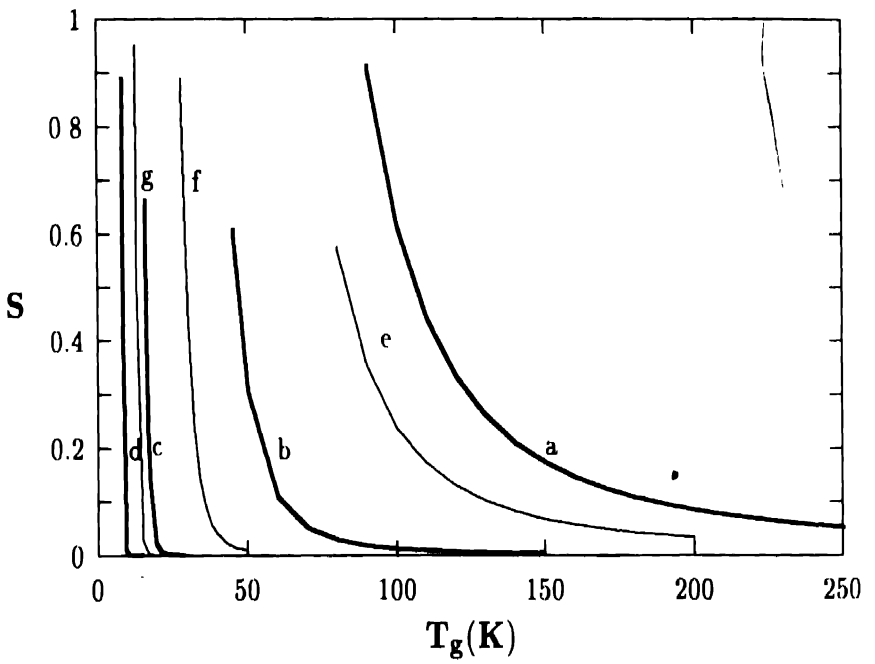
TABLE (2): Bound-state energies and transient times at a given temperature range.

systems	Substrate temp.range(K)	n	Theo.BS (K)	Expt.BS (K)	transient time(t_s)Sec.
He-LiF	34	0	62.69	68.47	7.87×10^{-9}
	08	1	32.16	28.55	2.17×10^{-8}
	02	2	11.72	9.05	8.27×10^{-7}
	01	3	1.38	2.44	1.50×10^{-5}
He-NaF	18	0	57.10	57.10	1.92×10^{-9}
	05	1	25.30	21.70	8.94×10^{-7}
	01	2	6.27	6.27	7.11×10^{-3}

The inverse of the imaginary part of the T-matrix gives the life time τ_S[4] and from the pole of the T-matrix we get BS energy and the corresponding surface temperature.

The figure shows the variation in sticking coefficient with gas temperature for He-LiF and He-NaF systems for different BS.Curves (a), (b), (c) and (d) are for He-LiF for n=0,1,2 and 3 respectively.Curves (e), (f), (g) are for He-NaF for n=0,1 and 2 respectively.

It is interesting to note from our tabulated results and the figure that at higher temperature the capture into the physisorbtion BS is possible. The measurement of such physorbed states however can be monitored through careful measurements of the specular beam.Thus we conclude that our theory which takes into account the higher order contribution in the transition matrix predicts a nonzero capture even at high temperature for a phonon mediated process.



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